LA-UR -91-2590

LA-UR--91-2590

DE91 017775

Received

SEP 0 6 1991

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

TITLE IDENTIFICATION OF ARTIFICIAL GAMMA-EMITTING NUCLIDES USING A SCINTILLATOR-BASED GAMMA-RAY SPECTRAL LOGGING SYSTEM

AUTHOR(S) J. G. Conaway, EES-3

MASTER

SUBMITTED TO 4th International MGLS/KEGS Symposium
Toronto, August 18-22, 1991

DISCLAIMER

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

FORM NO. 836 R4

IDENTIFICATION OF ARTIFICIAL GAMMA-EMITTING NUCLIDES USING A SCINTILLATOR-BASED GAMMA-RAY SPECTRAL LOGGING SYSTEM

John G. Conaway
Los Alamos National Laboratory

J. G. Conaway, Identification of artificial gamma-emitting nuclides using a scintillator-based gamma-ray spectral logging system; *in* Proceedings of the 4th International MGLS/KEGS Symposium on Borehole Geophysics for Minerals, Geotechnical and Groundwater Applications; Toronto, 18-22 August 1991.

ABSTRACT

The standard spectral gamma ray logging system used at the Nevada Test Site (NTS) contains a large sodium iodide detector which provides high efficiency but low energy resolution. To enhance the capabilities of this system for identifying artificial gamma-emitting nuclides, I developed and implemented a simple procedure for extracting artificial components from low-resolution gamma-ray spectra. This procedure uses three *basis* spectra, developed by a consultant using numerical modeling, representing the spectral response of the downhole instrument to naturally occurring potassium, uranium and thorium family gamma rays in a large-diameter air-filled borehole.

To extract the artificial spectral components, the three basis spectra are first scaled to the recorded field spectrum using the usual spectral windows for K, U and Th; these windows bracket the photopeaks at 1.46, 1.76 and 2.61 MeV. Since most of the contribution from artificial nuclides will fall below 1.26 MeV (the lower limit of the potassium window) this scaling process should be insensitive to the presence of artificial nuclides. The scaled basis spectra are then subtracted from the field data, leaving a *residual* spectrum consisting of noise plus the contribution of any artificial gamma-emitting nuclides. This process is repeated for each spectrum in the log, or the spectra can be accumulated over any desired depth range for better statistics. Rather than inspect each spectrum visually, a parameter can be computed which indicates the presence of artificial nuclides; this parameter can be plotted along with the usual K, U and Th concentration estimates as a function of depth. These techniques have been used successfully on field data and provide us with an inexpensive screening tool to detect artificial nuclides along boreholes.

INTRODUCTION

Although radionuclide migration is not thought to be a problem at the Nevada Test Site (NTS), it has come under increased scrutiny as have all environmental issues. Recently, a long-term study of this subject was begun involving a special drilling program along with geophysical logging, geological and hydrological studies, and other work. Clearly, it would be useful in such a study to have a logging device that could provide a reconnaissance scan of the borehole for artificial gamma-emitting nuclides.

Operational considerations aside, the instrument of choice for resolving characteristic gamma ray energies and identifying the emitting nuclides is a high-resolution gamma-ray spectral logging system based on a solid-state detector such as high-purity germanium. Such detectors provide high energy resolution at the expense of low detection efficiency, leading to very low logging speeds. Further, these are costly systems to develop, run and maintain. High resolution spectral logging systems are available as a commercial logging service, but the cost of running such a service routinely would be substantial.

At the NTS we have a low-resolution spectral gamma-ray logging system built to our specifications by the principal logging contractor there. This device contains a large sodium iodide detector (2150 cm^3 of NaI(Tl)) which provides high detection efficiency but low energy resolution. The system produces a spectrum of 256 channels which is transmitted digitally up the wireline. The commonly used windows approach for identifying potassium and gamma-emitting members of the uranium and thorium families is used by this contractor to process the data.

To improve the results from our scintillator based system, I developed a simple procedure for extracting the artificial components of low-resolution gamma-ray spectra. This procedure can be applied whenever the presence of artificial gamma-emitting nuclides in boreholes is known or suspected. The problem of detecting the presence of artificial nuclides against natural background gamma radiation has been discussed previously by Bristow (1978), Myrick et al (1983), Grasty and Multala (1991) and others.

THE BASIS SPECTRA

The key to the procedure used here to extract the artificial spectral components from recorded field spectra is a set of three *basis* spectra, one representing the ^{40}K spectrum and one each representing the equilibrium natural uranium and thorium decay series spectra (Figure 1). The basis spectra used in this study were developed for us by a consultant (see Acknowledgments) who performed

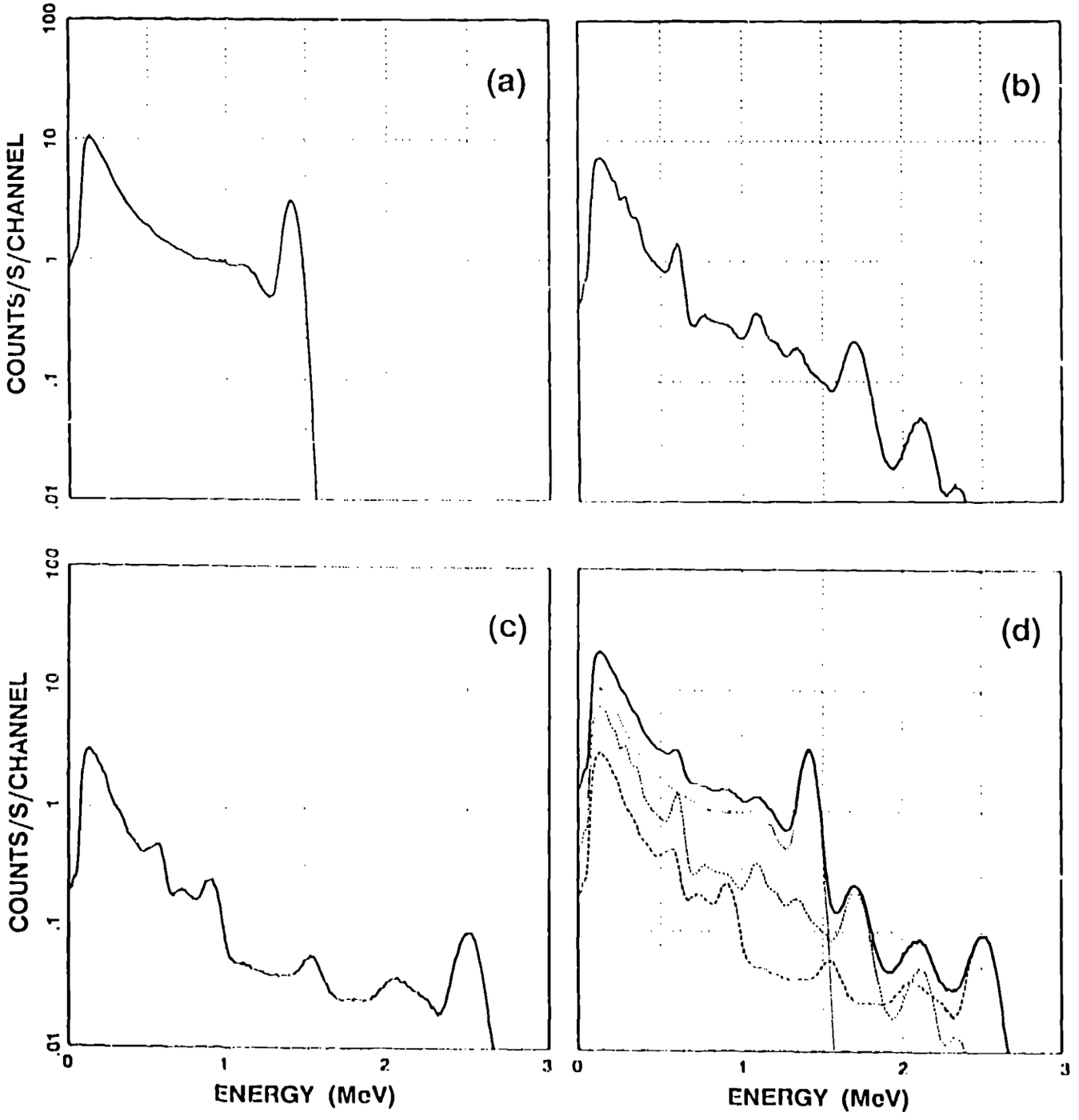

Figure 1 - The three basis spectra used in this study: (a) 1% potassium, (b) 1 ppm uranium, and (c) 1 ppm thorium. (d) The sum of the three individual spectra.

numerical simulations of the spectral gamma-ray logging system used at the Nevada Test Site under typical borehole conditions encountered there. The simulation model has been described by Wilson (1990) and Wilson and Conaway (1991).

The model spectra differ in certain minor ways from recorded spectra from the logging system. Recorded spectra suffer somewhat from differential non-linearity due to the successive-approximation analog-to-digital converter used in the instrument. This was kept small by the use of the 8 most significant bits from a 12-bit converter, but differential non-linearity effects are still visible in the recorded spectra. Statistical counting noise can be a problem in the recorded spectra when only small concentrations of radioelements are found in the formations, as at the Nevada Test Site. This has been overcome to some extent by the use of an unusually large detector for borehole applications. The field logs were run in a nominal 31 cm water-filled borehole while the basis spectra were developed assuming a 2.44 m diameter air-filled borehole. These basis spectra should be similar to those which would be observed in a 31 cm air-filled borehole since the attenuation due to air is small for gamma-ray energies of interest. The effect of the borehole water should also be relatively small, since the borehole sonde is some 20 cm in diameter and the water-filled annulus is an average of 5 cm across. For this initial study, I neglected the effect of the water on the shape of the basis spectra, although for best results the basis spectra should be recomputed for the appropriate borehole conditions.

Errors in the model spectra come from several sources. In particular, to reduce computation time the simulated source gamma-rays were lumped into 48 groups based on energy distribution and intensity (Wilson and Conaway, 1991). In some cases, several adjacent energy lines were grouped together into a single "peak"; care was taken not to smear major energy lines by this grouping process. As an additional problem, computation noise such as imperfect convergence and statistical noise affects the model spectra.

Even with the errors described above and other errors affecting the spectra, the fit between the recorded and model spectra was fairly good. This fit was refined by adjusting the energy resolution used in the model, by compensating the model spectra for a slightly nonlinear energy dependence of the recorded spectra, and by adjusting one of the 48 energy groups to improve the fit over one small energy region after careful comparison with field spectra. The final result was a set of basis spectra that proved to be quite adequate for the requirements of this study, although some additional improvement at lower energies would be useful.

THE RESIDUAL SPECTRUM

The approach used in this study is to subtract out the contributions of the naturally occurring radioelements from the recorded gamma-ray spectrum, leaving the contributions from artificial nuclides plus noise. This is fairly straightforward assuming the artificial component of the spectrum above 1.26 MeV is negligible, since the windows which we use to identify potassium (1.46 MeV), uranium (the 1.76 MeV line of ^{214}Bi from the ^{238}U decay series) and thorium (the 2.61 MeV line of ^{208}Tl from the ^{232}Th decay series) all fall above 1.26 MeV. This assumption proved to be valid for the data used in this study.

This is the procedure:

1. Read in a single spectrum or accumulate a spectrum over a specified depth interval.

2. Ascertain the position of the 2.61 MeV thorium peak to determine the system gain for that particular spectrum.

3. Re-channelize the spectrum so that offset is zero and gain is 10 keV per channel, so the gain and offset of the field spectrum correspond to the basis spectra.

4. Determine the relative contributions of gamma rays from potassium and from the uranium and thorium families to the spectrum at each depth point of the log. This was accomplished using the common energy window technique with spectral stripping.

5. Scale the three characteristic basis spectra by the radioelement concentrations determined in step 4.

6. Subtract the scaled basis spectra from the recorded spectrum to produce the residual spectrum.

This is repeated at each depth or for each desired depth range. The resulting *residual* spectrum at each depth is composed of any artificial spectral components plus noise. The noise consists of statistical fluctuations, instrumental noise, and differences between the shapes of the three characteristic spectra used and the actual shapes of those spectra which *should* have been used had they been known. These characteristic spectral shapes will vary somewhat with a number of factors such as borehole casing, diameter of a liquid-filled borehole, non-standard geometry (e.g. a layer of radioactive material on the borehole wall from the drilling fluid or ^{222}Rn gas infiltration), and will also vary with distance along the borehole away from a given zone of naturally radioactive rock (e.g. Czubek, 1961, 1969; Conaway, 1980; Wilson and Conaway, 1991).

THE ARTIFICIAL NUCLIDE PARAMETER

The residual spectrum described above can be further processed to obtain an artificial nuclide parameter (ANP) which can be plotted as a function of depth. I tried several parameters without any attempt at formal optimization, and found that the following parameter worked well for our application:

$$ANP = \frac{R_L - R_{II}}{S_{II}} \qquad (1)$$

where R_L is the countrate in a window at the lower energy end of the *residual* spectrum (from 0.4 to 1.0 MeV), R_{II} is the countrate in a window at the higher energy end of the *residual* spectrum (from 1.26 to 3.0 MeV) and S_{II} is the countrate in a window at the higher energy end of the *field* spectrum (from 1.26 to 3.0 MeV). 0.4 MeV was used as the lower summation limit for the lower energy window because the departure of the model spectra from field spectra increases at lower energies. Note that the ANP is a relative indicator; the absolute values have no useful meaning.

A FIELD TEST

The procedures described above were tested on data from a borehole for which a spectral gamma-ray log showed two anomalies in the total counts window which were not seen in the potassium, uranium or thorium windows (Figure 2). Under favorable conditions, artificial nuclides may be located, as they were in this case, by scanning the log for anomalies in the total counts curve which are not reflected in the radioelement curves. However, such a pattern might not stand out on a log in a borehole with a more complicated natural radioelement distribution. A plot of the ANP defined in Equation 1, or some other parameter of that type, gives a much better signal of the presence of artificial nuclides (Figure 3). Here, the anomalous zones stand out unambiguously from a nearly flat background.

A typical spectrum accumulated over a depth range from 2180 to 2210 feet (664.5 - 673.6 m), is shown in Figure 4a (note the logarithmic count rate axis.) This spectrum includes a number of identifiable photopeaks along with some instrumental and statistical noise. The three basis spectra are scaled and summed to provide the best fit to the field spectrum for energies over 1 MeV, shown along with the constituent (Figure 4b). Figure 4c shows the fit between field and model data, with the computer generated spectrum shown by the dotted curve. The composite spectrum is then subtracted from the field spectrum to produce the residual spectrum, as shown in Figure 4d (note the linear scale). Clearly, anything left in the residual spectrum is either the contribution of an artificial nuclide or (by definition) noise. In Figure 4d, the residual spectrum apparently consists entirely of noise; some sources of noise were discussed above.

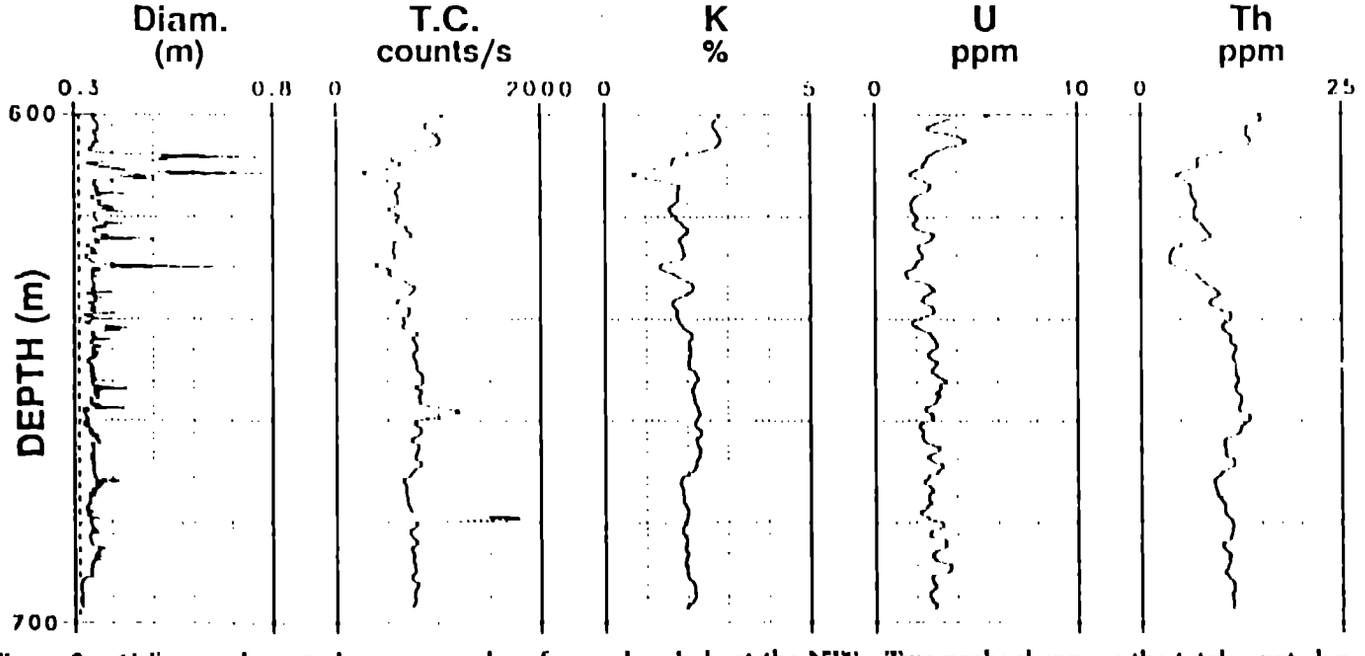

Figure 2 - Caliper and spectral gamma ray logs from a borehole at the NTS. Two peaks shown on the total counts log (T.C.) at about 658 m and 679 m are not seen on the K, U or Th curves and were therefore suspected of having been caused by artificial gamma-emitting nuclides along the borehole. Since this is a water-filled borehole, some of the larger washouts seen on the caliper log are reflected in the spectral curves as relative lows; no borehole size correction was applied to the data.

A second accumulated field spectrum is shown in Figure 5a, this one accumulated from 2225 to 2230 feet (678.2 - 679.7 m). The three basis spectra are again scaled and summed to provide the best fit to the field spectrum for energies over 1 MeV (Figure 5b and 5c). Even by eye we can see a distinct difference between these spectra at the lower energies. The composite spectrum is then subtracted from the field spectrum to produce the residual spectrum shown in Figure 5d. This time the residual spectrum clearly shows an artificial component, largely ^{137}Cs (0.661 MeV).

Another example of the effect of artificial nuclides is seen in the field data accumulated from 2156 to 2164 ft (657.1 - 659.6 m) shown in Figure 6. Again, the presence of ^{137}Cs is obvious after the natural spectral components have been removed (Figure 6d), at somewhat lower intensity than in the previous example.

THE POTENTIAL FOR QUANTITATIVE RESULTS

If quantitative estimates of artificial gamma-emitting nuclide concentrations are to be made based on a spectral gamma-ray log, some assumption must be made regarding the distribution of radioactive material. In the case of natural spectral gamma ray logging the assumption is generally made that the naturally occurring radioelements are uniformly distributed in layers of arbitrary thickness, and that the concentration may vary from layer to layer. If that assumption holds, then good estimates may be made of the concentrations (see, e.g., Scott, 1963; Czubek, 1961, 1971; Conaway et al, 1980). In the case of artificial nuclides the same is true if the nuclides are uniformly distributed in layers. However, it can be visualized that artificial nuclides may be present as a surface distribution on the borehole wall as a few discrete sources, distributed in the borehole fluid, or in

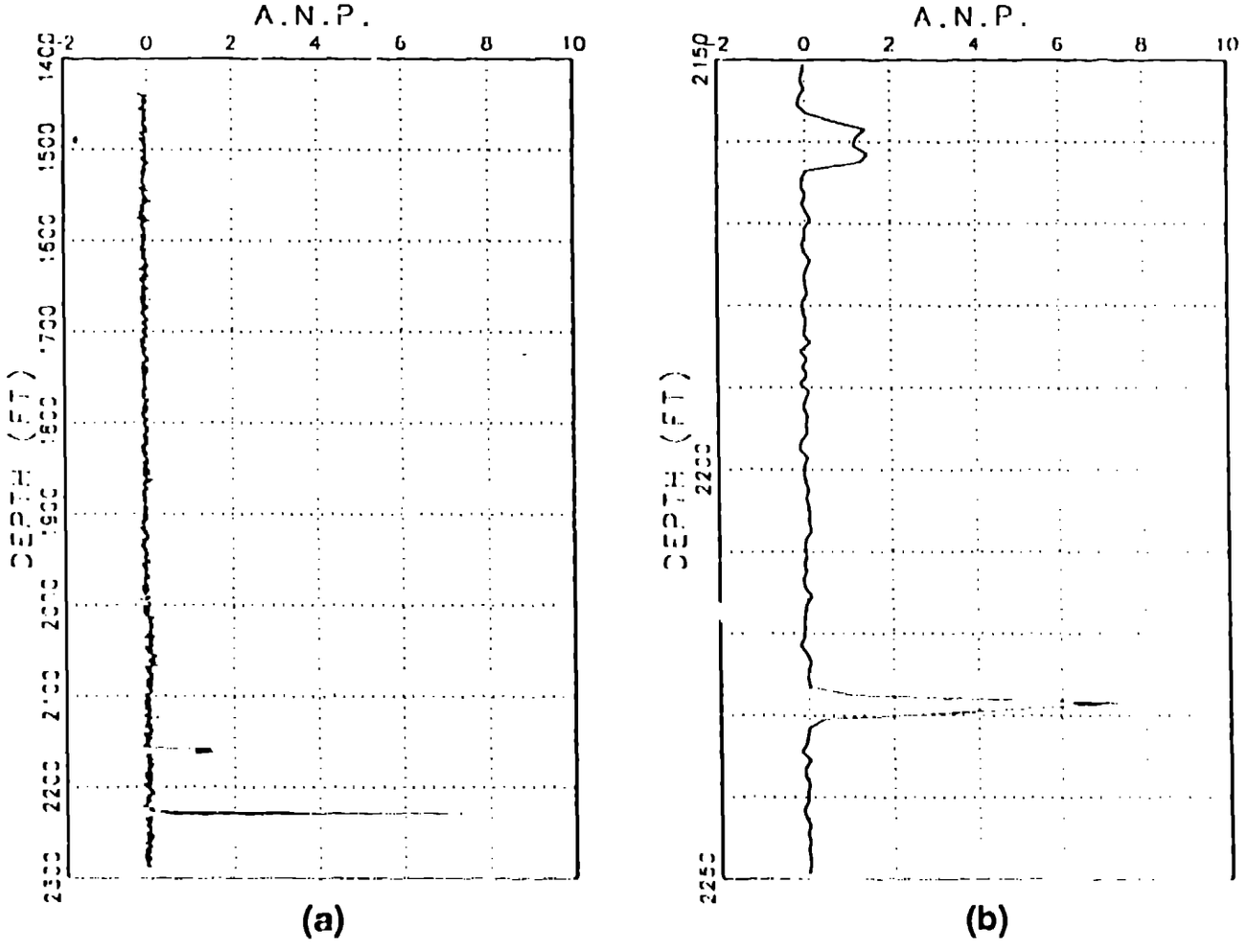

Figure 3 - (a) A plot of the Artificial Nuclide Parameter (ANP) computed by applying Equation 1 to the same gamma ray spectra used to produce the K, U and Th curves in Figure 2. (b) The same ANP data plotted at an expanded depth scale. The spectra were accumulated over 30 cm (1 ft) intervals; no smoothing was used in either curve.

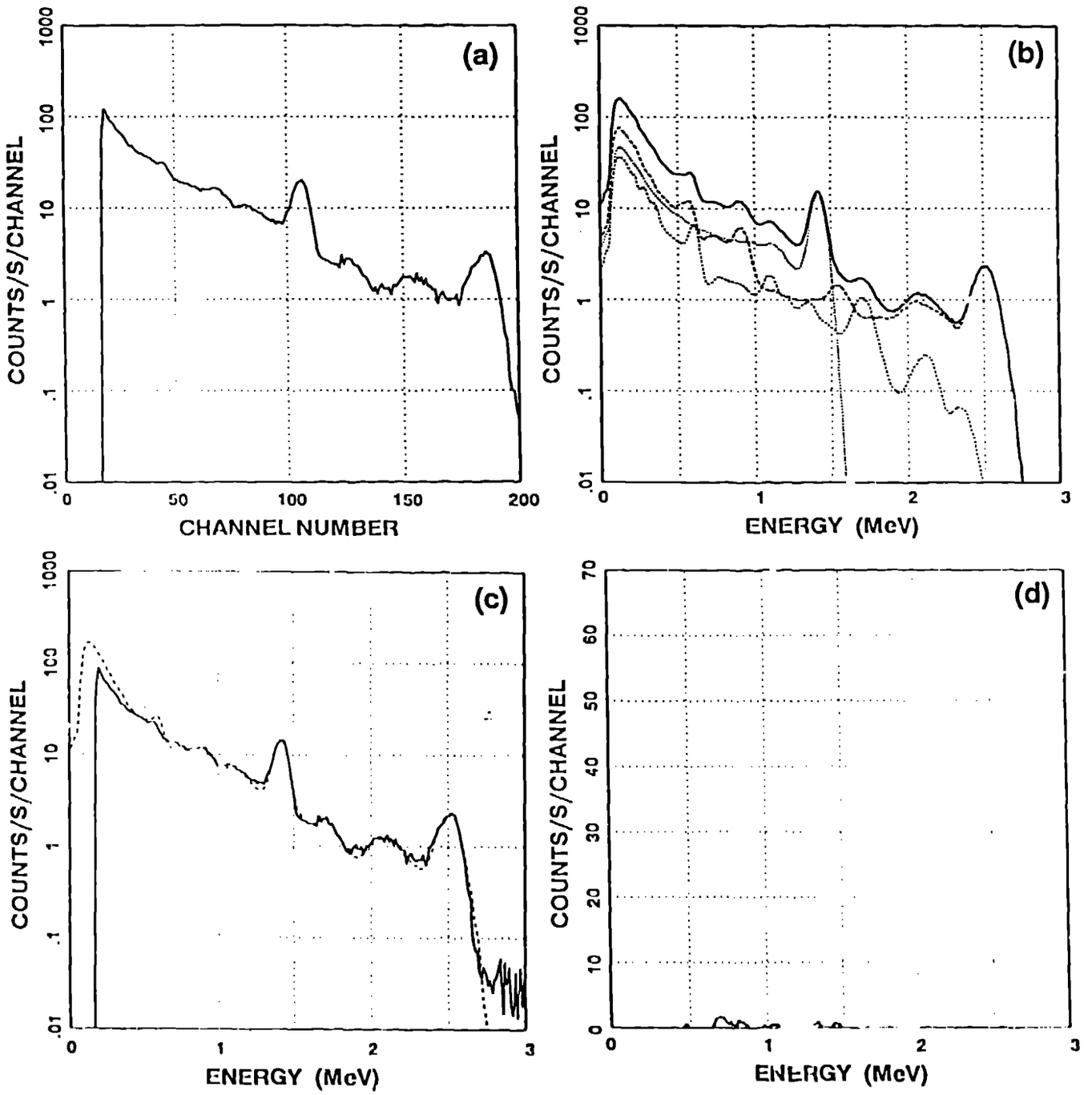

Figure 4 - (a) A field spectrum averaged over the depth range of 664.5 m to 673.6 m. (b) The corresponding scaled composite model spectrum (solid line) made up from the K, U and Th basis spectra (dashed lines). (c) The fit between the field spectrum (a) and the model spectrum (b). (d) The residual spectrum, computed by subtracting the model spectrum (b) from the field spectrum (a). In this case, no artificial spectral component is apparent.

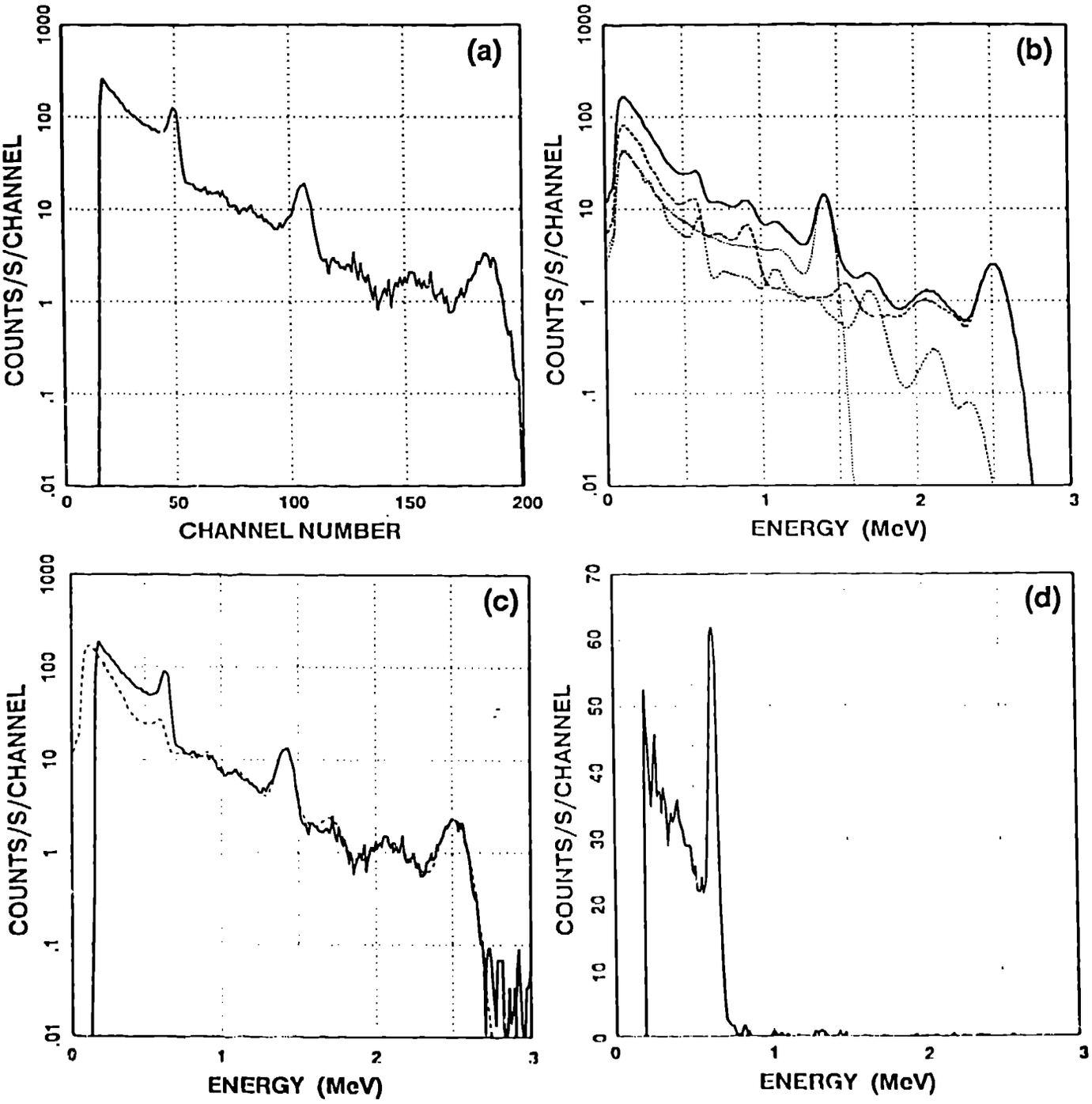

Figure 5 - (a) A field spectrum averaged over the depth range of 678.2 m to 679.7 m. (b) The corresponding scaled composite model spectrum (solid line) made up from the K, U and Th basis spectra (dashed lines). (c) The fit between the field spectrum (a) and the model spectrum (b). (d) The residual spectrum, computed by subtracting the model spectrum (b) from the field spectrum (a). In this case, an artificial spectral component is apparent, identified as ^{137}Cs based on the 0.661 MeV photopeak.

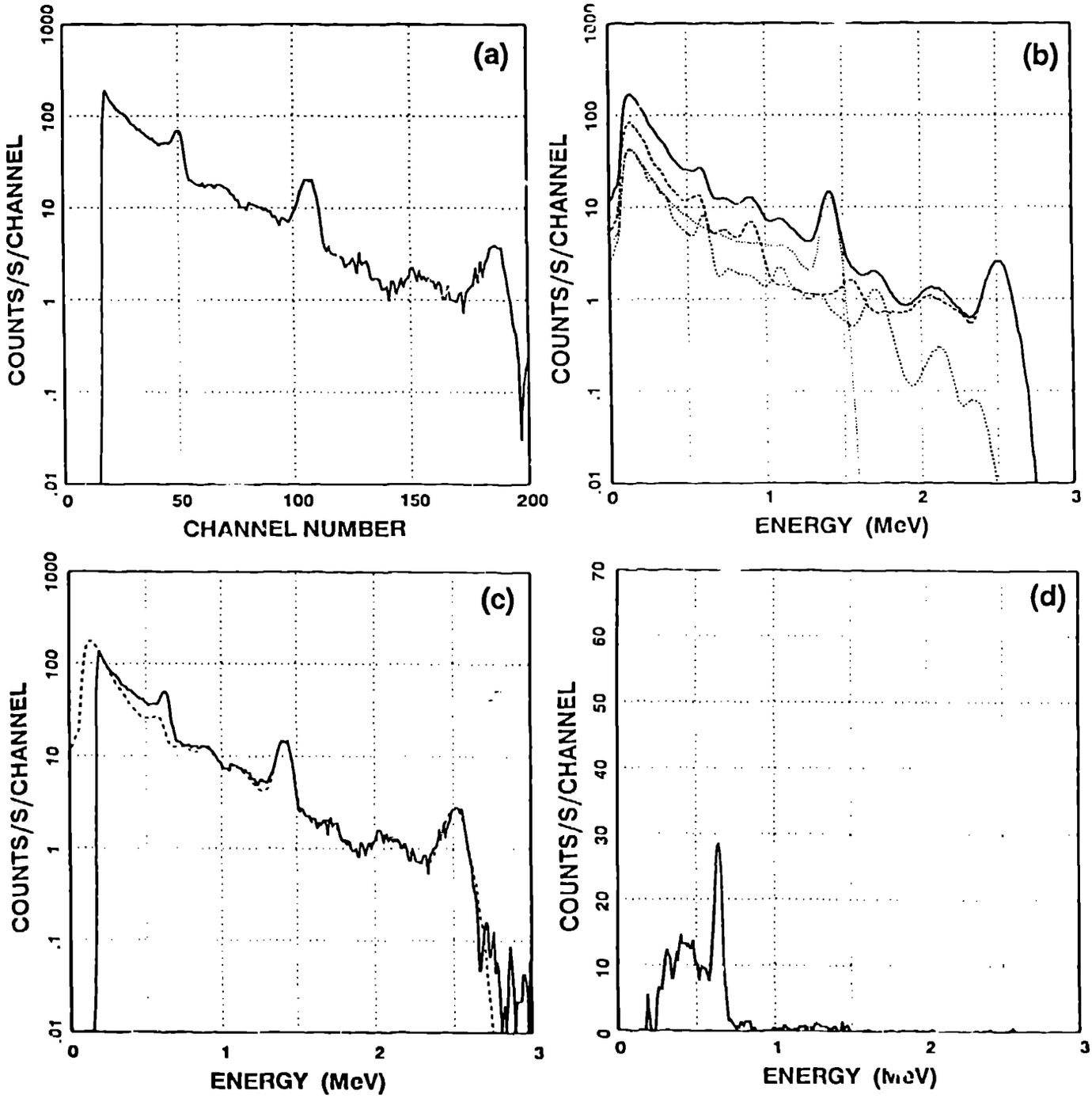

Figure 6 - (a) A field spectrum averaged over the depth range of 657.1 m to 659.6 m. (b) The corresponding scaled composite model spectrum (solid line) made up from the K, U and Th basis spectra (dashed lines). (c) The fit between the field spectrum (a) and the model spectrum (b). (d) The residual spectrum, computed by subtracting the model spectrum (b) from the field spectrum (a). As in Figure 5, ^{137}Cs is apparent, although at somewhat lower intensity than in the previous example.

some other geometry. With some of these geometries it should be possible to develop procedures for extracting quantitative estimates of radioelement concentrations, but the problem is not as straightforward as the layered-geology situation. An example of the additional complexities found in such situations was given by Wilson and Conaway (1991), where the problem of potassium-loaded mudcake was examined using computer modeling.

Under favorable conditions it should be possible to gain some idea of the distribution of the radioactive material, although probably not the actual quantity, based on the shape of the residual spectrum since, for instance, the spectrum resulting from a surface source is harder (i.e. contains fewer scattered gamma rays) than the spectrum from a source distributed through the rock. Developing such an approach is theoretically possible if the noise level of the data is low enough. In this study of ^{137}Cs in a borehole at the NTS, no effort was made to obtain quantitative results.

DISCUSSION AND CONCLUSIONS

^{137}Cs is a common constituent of man-made radioactive contamination but, since the 0.661 MeV photopeak of ^{137}Cs overlaps two major natural photopeaks (0.609 MeV from the U series and 0.581 MeV from the Th series), it may not be easy to look at a field spectrum, such as the ones shown in Figures 4a, 5a and 6a, and tell if contamination is present unless the contribution of the artificial component is overpowering. Subtracting channel by channel an appropriate computer-generated spectrum representing the contributions of K, U and Th greatly enhances our ability to identify artificial nuclides.

Although the method given in this paper for identifying gamma-emitting contaminants may not be the equal of using a high-resolution spectral gamma-ray logging system based on a solid state detector, it nonetheless provides an efficient on-site capability for reconnaissance. Using our existing spectral gamma ray logging system along with the data processing procedure presented above as a reconnaissance scan for artificial nuclides represents a substantial cost savings compared with using a commercial high-resolution spectral logging service.

ACKNOWLEDGMENTS

This work was funded by the United States Department of Energy. The basis spectra used in this study were developed by Dr. Robert D. Wilson and his colleagues at Highland Scientific, Ltd. (Ashland, Oregon), under contract to Fenix and Scisson of Nevada, a USDOE NVOO contractor.

REFERENCES

Bristow, Q., 1978: The application of airborne gamma-ray spectrometry in the search for radioactive debris from the Russian satellite Cosmos 954 (Operation "Morning Light"); *in* Current Research, Geol. Surv. Can. Paper 78-1B, p. 151-162.

Conaway, J.G., 1980: Uranium concentrations and the system response function in gamma ray logging; *in* Current Research, Part A, Geological Survey of Canada Paper 80-A, p. 77-87.

Conaway, J.G., Bristow, Q., and Killeen, P. G., 1980: Optimization of gamma-ray logging techniques for uranium; Geophysics, V. 45, p. 292-311.

Czubek, J.A., 1961: Some problems of the theory and quantitative interpretation of the gamma ray logs; Acta Geophysica Polonica, V. 9, p. 121-137.

Czubek, J.A., 1969: Influence of borehole construction on the results of spectral gamma logging: *in* Nuclear Techniques and Mineral Resources, International Atomic Energy Agency Proceedings Series, IAEA, Vienna.

Czubek, J.A., 1971: Differential interpretation of gamma-ray logs: I - Case of the static gamma-ray curve: Rep. no. 760/I, Nuc. Ener. Inf. Center, Polish. Gov. Comm. for Use of Nuclear Energy, Warsaw, Poland.

Grasty, R.L., and Multala, J., 1991: A correlation technique for separating natural and man-made airborne gamma-ray spectra; *in* Current Research, Part D, Geological Survey of Canada, Paper 90-1D, p. 111-116.

Myrick, T. E., Blair, M. S., Doane, R. W., and Goldsmith, W. A., 1983: A mobile gamma scanning system for detecting radiation anomalies, Nuclear Technology, V. 62, p. 364-370.

Scott, J. H., 1963: Computer analysis of gamma-ray logs; Geophysics, V. 26, p. 457-465.

Wilson, R. D., 1990: Simulations of the Nevada Test Site spectral gamma-ray logging tool; Final Report submitted to Fenix and Scisson of Nevada, Corp., 103 pp.

Wilson, R. D., and Conaway, J. G., 1991: Radiation transport simulations of a spectral gamma-ray logging tool in the presence of a surface distribution of radioactive material on the borehole wall. IEEE 1991 Nuclear Science Symposium, Nov. 5-9, 1991, Santa Fe.